Dynamical nuclear spin polarization and the Zamboni effect in gated double quantum dots

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A dynamical nuclear polarization scheme is studied in gated double dots. We demonstrate that a small polarization ($\sim 0.5\%$) is sufficient to enhance the singlet decay time by two orders of magnitude. This enhancement is attributed to an equilibration process between the nuclear reservoirs in the two dots accompanied by reduced fluctuations in the Overhauser fields, that are mediated by the electron-nuclear spin hyperfine interaction.

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Electron spins localized in semiconductor quantum dots have been intensively investigated in recent years due to their potential use in quantum information processing and spintronics.[1] Several experimental[2,3,4] and theoretical[5,6,7,8,9,10] studies have identified the hyperfine (HF) interaction between an electron spin and the surrounding nuclear spins as one of the main sources for electron spin decoherence in low temperature GaAs quantum dots, leading to T_2^* on the order of $10-25$ns and T_2 on the order of μs. These values are orders of magnitude shorter than the spin relaxation time, which approaches tens of milliseconds in these systems.[2]

Several strategies have been suggested to alleviate electron spin decoherence via nuclear spins, including spin-echo techniques (to remove inhomogeneous broadening[4,7,10]), nuclear spin state measurement (to narrow the Overhauser field distribution,[11]) and nuclear polarization (to reduce phase space for nuclear spin dynamics.[12]) Nuclear spin polarization is also valuable for state initialization in NMR quantum computing and for utilizing collective nuclear states as long-lived quantum memory.[13] So far, optical pumping has produced up to $\sim 60\%$ nuclear polarization[14] in interface fluctuation GaAs dots, while spin transfer via hyperfine mediated spin-flip scattering in the spin-blockade regime in gated GaAs dots has led to $\sim 1\%$ polarization.[15,16] Among the limiting factors in these dynamical polarization schemes are the large difference in the Zeeman energies of the electrons and nuclei making joint spin flip processes energetically unfavorable, and nuclear spin diffusion due to dipolar interaction. In contrast, theoretical studies have shown that in order to achieve a sizeable enhancement of single electron spin decoherence time via phase-space squeezing, a nuclear polarization of more than 99% is required.[8,9,17]

In this paper we show that HF interaction can be exploited to dynamically polarize the nuclear spins in gated double dots. Most interestingly the relaxation time of the two-electron spin singlet state dramatically increases without the need for nearly complete nuclear polarization. This suppression of relaxation is achieved by an equilibration process in the nuclear reservoirs in the two dots and a reduction in the fluctuations of their Overhauser fields. We have dubbed this effect, mediated by the HF interaction during the polarization cycles, as the nuclear Zamboni effect.[18]

We study the dynamics of a system of two electrons localized in a gated double dot interacting with two nuclear spin baths within the framework of the Hamiltonian

$$H = H_{\rm orb} + H_Z + H_{\rm HF}, \qquad (1)$$

where we have neglected nuclear-nuclear dipolar coupling in the current study. For the orbital part we adapt the Hund-Mulliken approach[12,19] to solve for the electronic states in the gated dot configuration, where $H_{\rm orb}$ includes the single-particle Hamiltonian and the Coulomb interaction. The relevant Hilbert space of $H_{\rm orb}$ is spanned by four two-particle states, $\{S(2,0), S(0,2), S(1,1), T(1,1)\}$, which consist of the separated singlet and triplet and the two doubly occupied singlet states. Indices (i,j) indicate the number of confined electrons in the (left,right) dot. We neglect the doubly occupied triplet states as their energy is much higher for the structures under study.[2,4] The Zeeman interaction $H_Z = g\mu_B \mathbf{B}\cdot\sum_{i=L,R}\mathbf{S}_i$, with $g=-0.44$ and μ_B being the Bohr magneton, splits the triplet states. The resulting energy diagram and exchange interaction J near the (1,1) to (0,2) charge transition are shown in Figs. 1a,b for $B=100$ mT ($E_Z = 2.5\mu$eV), half interdot distance $a = 1.9a_B$, and dot confinement $\omega_0 = 120\mu$eV, corresponding to the experimental parameters in ref. 4.

For the HF interaction we consider the contact term between the two electrons and the surrounding nuclei

$$H_{\rm HF} = \sum_{i=L,R}\sum_k A_i^k \mathbf{I}_i^k \cdot \mathbf{S}_i = \mathbf{h}\cdot\mathbf{S} + \boldsymbol{\delta}\mathbf{h}\cdot\boldsymbol{\delta}\mathbf{S}\,, \qquad (2)$$

where A_i^k is the HF coupling constant with the kth nucleus in the ith dot. Here $\mathbf{h} = \frac{1}{2}(\mathbf{h}_L + \mathbf{h}_R)$, $\boldsymbol{\delta}\mathbf{h} = \frac{1}{2}(\mathbf{h}_L - \mathbf{h}_R)$ and $\mathbf{S} = \mathbf{S}_L + \mathbf{S}_R$, $\boldsymbol{\delta}\mathbf{S} = \mathbf{S}_L - \mathbf{S}_R$ are the sums and differences of the nuclear fields and the electron spins in the two dots, respectively, where $\mathbf{h}_i = \sum_k A_i^k \mathbf{I}_i^k$ is the nuclear field in dot i. We assume $I_i^k = \frac{1}{2}$ for simplicity. The Hamiltonian in Eq. (1) conserves the total spin and can be block diagonalized in each of the Hilbert subspaces defined by the eigenvalues of the operator $J_z = S_L^z + S_R^z + \sum_{i,k} I_i^{kz}$. In order to make the numerical effort for a larger number of spins tractable, we assume a constant HF coupling for all the nuclei in each dot, allowing us to write the Overhauser fields as $\mathbf{h}_i = (\gamma_i/N)\mathbf{I}_i$, $i = L, R$, where $\gamma_i = \sum_k A_i^k \approx 100$ μeV is the total HF coupling, N is the number of nuclei per dot, and $\mathbf{I}_i$ is the collective spin operator for dot i. This approximation provides two more integrals of motion, namely the two $SU(2)$ Casimir operators $\mathbf{I}_L^2$, $\mathbf{I}_R^2$, and enables us to further divide the Hilbert space, making the complexity of the problem scale polynomially with N instead of exponentially.

We have tested the validity of the uniform HF coupling approximation for the nuclear polarization dynamics studied here by slicing the dot into concentric rings. Assigning

different HF coupling for each ring, and assuming no inter-ring dynamics, we find that the dynamics described below are largely unaffected by this averaging procedure.[20] Studies of nuclear spin diffusion in a quantum dot also verify that inter-ring nuclear spin dynamics is significantly suppressed for smaller quantum dots.[21] Our approach enables us to study the interplay between HF and exchange effects within a unified theory through exact diagonalization of the Hamiltonian (1). The system dynamics under any applied gate-pulse are calculated without resorting to the quasi-static approximation that may not be appropriate to describe nuclear polarization dynamics.[22] At the same time, our use of collective spin states enables us to consider a substantially larger number of nuclei (~ 1000 per dot) as compared with previous numerical studies,[6,7] which is important in determining the scaling properties of the dynamical features with N.

The nuclear spin configuration in each dot is represented in the basis of collective Dicke states $|j,m\rangle$, where $0 \le j \le N/2$ is the total spin of the state (or cooperative number) and $|m| \le j$ is the z projection of the total spin. The initial nuclear spin configuration in the two dots is written as

$$|\psi(0)\rangle_{\rm nuc} = \sum_{I_L=0}^{N/2} w(I_L)|I_L, I_L^{z0}\rangle \otimes \sum_{I_R=0}^{N/2} w(I_R)|I_R, I_R^{z0}\rangle, \quad (3)$$

and will be denoted henceforth as $(I_L^{z0}, I_R^{z0})_w$ where I_i^{z0} indicates the initial polarization of the nuclear configuration in dot i. The distribution weights $w(I_i)$ are assigned statistically by the number of possible ways in which one can arrange N spins into singlets, triplets, quintets, etc. We find that these weights obey a Gaussian distribution peaked at $I_i = \sqrt{N/2}$ whose width is $\sqrt{2N}$, as shown in Fig. 1c.[23]

An example of the proposed dynamical polarization pulse sequence is shown in the lower panel of Fig. 1a.[16] The key is to drive the system through the $S-T_+$ resonance with different speed in the two directions, so that only in one direction can the electron-nuclear spin flip occur. An $S(0,2)$ state is prepared by positively detuning the double dot (P) to enabe electron exchange with the leads, whose Fermi level is above the $S(0,2)$ but below the doubly occupied triplets. The electrons are then separated using rapid adiabatic passage, where the bias is swept to a negative detuning quickly relative to the HF coupling but slowly as compared to the electron tunneling between the hybridized singlet states. Our simulations show that the adiabaticity requirement is met with sweep times of 5 ns. The bias is then swept back slowly through the $S-T_+$ anticrossing (D, which can be identified experimentally[4]) where the HF interaction mediates electron spin flip flop with the nuclear spin baths. Choosing point S' to be far enough from the $S-T_0$ degeneracy, the electron spin state is always flipped from a singlet to a T_+, thus polarizing the nuclear spin baths. Finally, the system is swept back to point P where the triplet state relaxes quickly through electron exchange with the leads, and a new $S(0,2)$ state is prepared for the next cycle. The singlet preparation at the end of each cycle is simulated by partially tracing the electronic subsystem and applying a direct product of the resulting nuclear density matrix with the electronic configuration at point P. To study effects of the polarization cycles on the relaxation of the electron singlet state, we add a measurement cycle where the dots are negatively detuned to the $S-T_0$ degeneracy (S) for a separation time τ_S, followed by a measurement of the singlet probability P_S ($\tau_M \sim 5\mu$s).[2,4] To enhance the efficiency of the polarization process, we perform a nonlinear bias sweep, spending a substantial part of the cycle in the vicinity of D (see Fig. 1a).

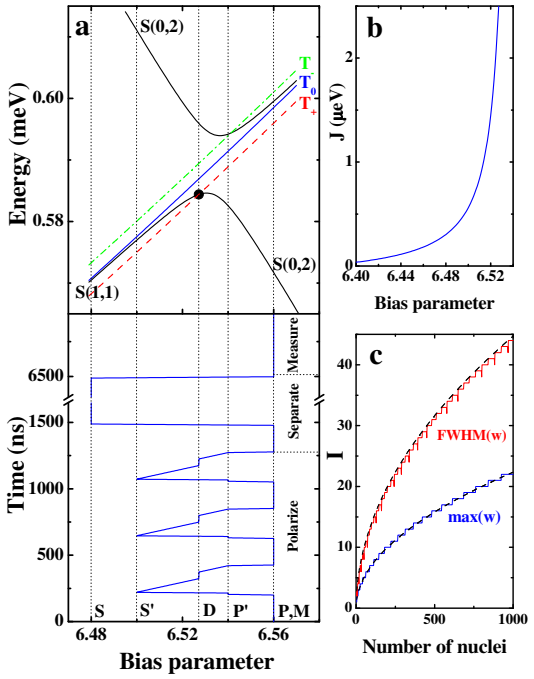

FIG. 1: (Color online) (a) Upper panel: Orbital energy diagram for the double dot near the (1,1)-(0,2) transition vs. a bias parameter, proportional to the inter-dot bias gate potential. Shown are the hybridized singlet states (black curves), and split (1,1) triplet states T_- (dash-dotted green), T_0 (blue), and T_+ (dashed red). • denotes the $S-T_+$ degeneracy point. Lower panel: Polarize, separate and measure pulse sequence. Three polarization cycles are shown followed by a separation time and a measurement of the singlet probability. The letters at the bottom indicate biasing points discussed in the main text. (b) Exchange energy as a function of bias. (c) Center location (blue) and FWHM (red) of the nuclear spin state distribution as a function of N. The dashed lines are $\sqrt{N/2}$ and $\sqrt{2N}$, corresponding to the center location and FWHM of the distribution, respectively.

Before presenting the simulation results for the polarization scheme, we discuss the envisaged impact of this procedure on the decay time of the electron singlet correlations. Figure 2 depicts the time evolution of P_S when the electrons are prepared in a singlet state and placed at point S, for several values of N. In Fig. 2a the initial nuclear state is $(0,-\min[\sqrt{N},I_R])_w$, representing a statistical polarization difference between the two dots. The time axes are multiplied by $\sqrt{N/10^5}$, indicating that the decay time scales like $1/\sqrt{N}$. The decay time of 25 ns agrees well with the experimental findings,[4,24] and its scaling with N corresponds to the decoherence time behavior found in ref. 8, since we scale the HF coupling constant with a fixed number of nuclei (10^5) rather then with N. Sim-

ilar dynamics is found for other initial nuclear states, e.g., $(-\min[\sqrt{N}, I_L], -\min[2\sqrt{N}, I_R])_w$. Fig. 2b shows the time evolution of P_S for the fully polarized nuclear configuration $(-I_L, -I_R)_w$. In this case P_S decay times are two orders of magnitude longer reaching 3μs and they do not scale with N. Other equally polarized configurations do not always present an enhanced singlet coherence, indicating that the fully polarized nuclear state is characterized by a narrower distribution in addition to having $\delta h_z = 0$. We stress that the term fully polarized does not suggest that all the spins are polarized, since one is limited by the total spin of each collective state within the weighted distribution. In fact, the total attainable polarization is $p = 1.4533/\sqrt{N}$ reaching a value of 0.46% for $N = 10^5$, which is consistent with recent experiments.[16,25]

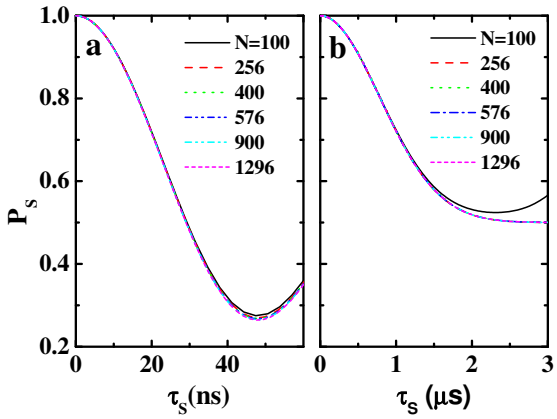

FIG. 2: (Color online) Electron singlet probability as a function of separation time for N values ranging from 100 to 1296, and $J = 0$. (a) Initial nuclear configuration $(0, -\min[\sqrt{N}, I_R])_w$. Time axes are scaled like $\sqrt{N/10^5}$, so that the presented decay times correspond to $N = 10^5$. The rise in P_S indicates decayed oscillations, which are an artifact of the uniform coupling approximation and can be eliminated by performing dynamics averaging over different HF couplings. (b) Initial nuclear configuration $(-I_L, -I_R)_w$.

The short time dynamics can be understood within a 2×2 effective Hamiltonian for the $S_z = 0$ subspace, $H_{\rm eff} = \frac{J}{2}(1 + \tau_z) + \delta h_z \tau_x$, where $\boldsymbol{\tau}$ is the pseudospin operator ($|S\rangle \rightarrow |\tau_z = -1\rangle$, $|T_0\rangle \rightarrow |\tau_z = 1\rangle$).[8] Even when $\langle \delta h_z \rangle = 0$ as for the fully polarized state in Fig. 2b, the spin dynamics do not vanish altogether. The reason is that applying $H_{\rm eff}$ on each of the collective states in the weighted distribution results in different eigenvalues and their dynamics do not cancel out, giving rise to quantum fluctuations. In both cases shown in Fig. 2 there is a long time ($\sim 2.5\mu$s for $B = 100$ mT) envelope decay attributed to higher-order corrections to $H_{\rm eff}$ that are contained in Eq. (1). This envelope scales like $1/N$ and could therefore govern the dynamics of the fully polarized state in the large N limit. Its long timescale is a consequence of the large Zeeman splitting as compared with the Overhauser fields and it can be made longer using a larger magnetic field. For the N values we are considering, these corrections are only observed for the state $(0, 0)_w$ for which all other dynamics are shut down.

Our results also agree well with the experimental findings[24] and analytical results[8] for $J > 0$. These include preservation of the singlet correlations over a long timescale in the limit of $J \gg E_{\rm nuc}$, and the appearance of damped oscillations in P_S with a saturation value that depends on $E_{\rm nuc}/J$ in the intermediate regime $J \sim E_{\rm nuc}$. We also find the long time ($\tau_S \gg T_2^*$) value of P_S to be $1/2$ for $B \gg B_{\rm nuc}$ and $1/3$ for $B \ll B_{\rm nuc}$, in agreement with semiclassical theory[5].

Now we investigate the effects of the polarization cycles on the electron spin states by separating the electrons every four cycles to calculate P_S as the polarization progresses. Bias changes require calculating the evolution separately for each bias, using the resulting density matrix at each step as the initial condition for the subsequent step. The numerical effort is thus much more demanding and we are limited to several tens of nuclei per dot. Figs. 3a-c show P_S calculated for the initial nuclear configuration, after 20 polarization cycles, and after 100 cycles. The singlet decay times show a gradual enhancement as the nuclear polarization builds up, and their scaling with N gradually shifts from $1/\sqrt{N}$ (Fig. 3a) to 1 (Fig. 3c). An enhancement of factor 300 is obtained for the singlet decay times that reach $\sim 8\mu$s when the polarization process is complete. The corresponding nuclear polarizations in the two dots, shown in Fig. 3d, equilibrate during the polarization process. This equilibration effect is robust to any of our choices of initial nuclear configuration, and the degree of equilibration depends on the symmetry of the double dot. The equilibration between the two nuclear spin configurations and the narrow distribution of the Overhauser fields formed during the polarization process are responsible for the prolonged singlet decay time.

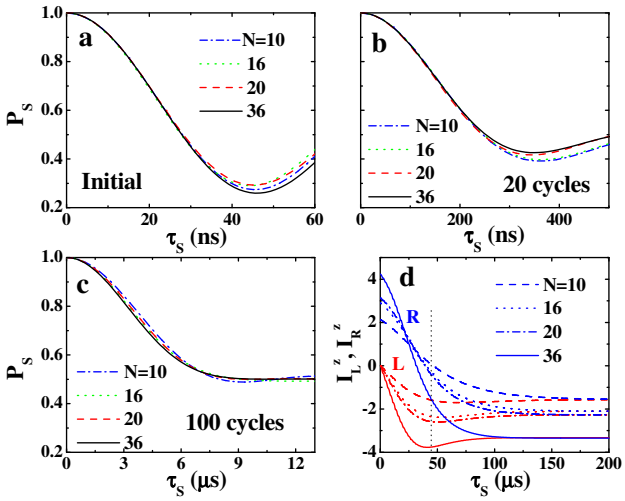

FIG. 3: (Color online) Singlet probability vs. separation time, calculated after performing polarization cycles, for several N values. The initial nuclear configuration is $(0, -\min[\sqrt{N}, I_R])_w$ and $J = 0$. (a) Initial P_S without polarization. Time axes are scaled like $\sqrt{N/10^5}$ (b) P_S calculated after 20 polarization cycles. Time axes are scaled like $(N/10^5)^{0.28}$ (c) P_S calculated after 100 polarization cycles. Time axes do not scale with N (d) The corresponding nuclear polarizations in the left (L-red lines) and right (R-blue lines) dot. The vertical dotted line corresponds to the time elapsed after 20 cycles, at which P_S shown in (b) was calculated.

Dipolar interaction between the nuclear spins can break

the weighted distribution of the spin collective states, and compete with the equilibration process. The number of polarization cycles needed to complete the process is $n_{\rm cyc} = 2\sum_{I=0}^{N/2} w_I^2 I \approx 1.4533\sqrt{N}$. The spin transfer time at point D is found to be $15.5\mu{\rm s}/\sqrt{N}$. As long as the $S-T_+$ degeneracy can be determined accurately, much of the cycle time in the large N limit is spent to prepare a singlet at P. Taking $t_P = 200$ ns for the electron exchange with the leads, the resulting time to complete the polarization is $\sim 120\mu$s.

A simple phase space argument seems to suggest that the interdot Zamboni effect should occur naturally through nuclear spin exchange between the two baths mediated, e.g., by one or two electrons in the double dot. While the fully polarized state has a single configuration, the non-polarized state is highly degenerate having $C_N^{N/2}$ configurations (assuming equal HF coupling for all the nuclei). Even for states with statistically similar dot polarizations the phase space difference could drive an equilibrating process. For example, the ratio between the number of configurations in $(0,\sqrt{N})_w$ and $(\sqrt{N}/2,\sqrt{N}/2)_w$ is $e^{-1/4}$ which, as our simulations show, is sufficient to induce an equilibrating process. We have performed simulations for the singlet decay times after a nuclear preparation time $\tau_{\rm eq}$ (on the order of tens of μs) in which we introduce one or two electrons into the double dot with appropriate bias and magnetic field. Interestingly, in spite of the equilibration of the two nuclear configurations, the singlet decay times are only modestly extended. This is because the distribution over I_i^z of the collective nuclear states is broadened during these naturally occurring electron-mediated equilibration processes. The time scale for this nuclear dynamics is in the order of $hNE_Z/\gamma_i^2 \approx 20$ ns, comparable to that for the electron spin dynamics, so that quasi-static approximation for the nuclear quantities becomes invalid in the regime of $\langle\delta h_z\rangle \approx 0$. The resulting fluctuations suppress the singlet decay time enhancement even though δh_z becomes small. In contrast, such dynamics does not exist in the fully polarized collective states, where the pumping of singlet states pushes I_i^z to their minimum values. The polarization cycles, while unable to produce high degrees of polarization, significantly regulate the nuclear spin states and reduce the nuclear field fluctuations, thus extend the decay time of the singlet state.

In summary, we have studied a nuclear polarization scheme in gated double dots utilizing the $S-T_+$ degeneracy point, and examined its impact on the two-electron spin singlet decay time, obtaining two-orders-of-magnitude enhancement. We have shown that high degree of nuclear polarization is not essential to suppress the nuclear relaxation channel for a two-electron spin state. Instead, enhancement of the singlet relaxation time is obtained by electron-mediated equilibration process within the two nuclear baths that suppresses the Overhauser field fluctuations within each reservoir. We have explored other strategies to facilitate this equilibrating process, and have found that while equal nuclear polarizations between the two dots may be obtained, they are not accompanied by narrowing of the nuclear state distribution and thus do not result in a dramatic enhancement in the singlet decay time.

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[1] I. Zutic, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[2] A. C. Johnson *et al.*, Nature **435**, 925 (2005); R. Hanson *et al.*, Phys. Rev. Lett. **94**, 196802 (2005).

[3] F. H. L. Koppens *et al.*, Science **309**, 1346 (2005).

[4] J. R. Petta *et al.*, Science **309**, 2180 (2005).

[5] I. A. Merkulov, Al. L. Efros, and M. Rosen Phys. Rev. B **65**, 205309 (2002).

[6] J. Schliemann, A. V. Khaetskii, and D. Loss, Phys. Rev. B **66**, 245303 (2002).

[7] N. Shenvi, R. de Sousa, and K. B. Whaley, Phys. Rev. B **71**, 224411 (2005).

[8] W. A. Coish and Daniel Loss, Phys. Rev. B **72**, 125337 (2005).

[9] C. Deng and X. Hu, Phys. Rev. B **73**, 241303(R) (2006).

[10] W. Yao, R. B. Liu, and L. J. Sham, Phys. Rev. B **74**, 195301 (2006).

[11] D. Klauser, W. A. Coish, and Daniel Loss, Phys. Rev. B **73**, 205302 (2006); D. Stepanenko, G. Burkard, G. Giedke, and A. Imamoglu, Phys. Rev. Lett. **96**, 136401 (2006).

[12] G. Burkard, D. Loss, and D. P. DiVincenzo, Phys. Rev. B **59**, 2070 (1999).

[13] J. M. Taylor, C. M. Marcus, and M. D. Lukin, Phys. Rev. Lett. **90**, 206803 (2003).

[14] A. S. Bracker *et al.*, Phus. Rev. Lett. **94**, 047402 (2005).

[15] K. Ono and S. Tarucha, Phys. Rev. Lett. **92**, 256803 (2004).

[16] J. R. Petta and C. M. Marcus (unpublished).

[17] R. de Sousa and S. Das Sarma, Phys. Rev. B **68**, 115322 (2003).

[18] Zamboni Machine, named after its inventor Frank J. Zamboni, has become a generic name for resurfacing ice rink machines. The analogy to the supression of quantum fluctuations in the nuclear reservoirs was suggested by C. M. Marcus.

[19] X. Hu and S. Das Sarma, Phys. Rev. A **61**, 062301 (2000).

[20] Inter-ring dynamics would result in the center rings being polarized first. The outer rings, for which the nuclear dynamics is slower, will tend to depolarize the nuclei in the center. Nevertheless, once the polarization is complete, all of the rings' collective nuclear states will be polarized, resulting in a qualitatively similar distribution to the one that is considered in the main text. Further study is needed to quantify the effects of inter-ring dynamics.

[21] C. Deng and X. Hu, Phys. Rev. B **72**, 165333 (2005).

[22] J. M. Taylor, J. R. Petta, A. C. Johnson, A. Yacoby, C. M. Marcus, and M. D. Lukin, cond-mat/0602470 (unpublished).

[23] This nuclear state distribution is in fact wider than the distribution obtained when considering non-uniform HF couplings, the latter given by a sum of Gaussian distributions corresponding to the number of nuclei in each ring.

[24] E. A. Laird *et al.*, Phys. Rev. Lett. **97** 056801 (2006).

[25] Slicing the dot to 100 rings gives $p = 4.3\%$ for $N = 10^5$. The HF coupling differences within each ring are then smaller than 0.1%, roughly preserving the total spin numbers in each ring.